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Transient lasing without inversion

Anatoly A Svidzinsky^{1,2,4}, Luqi Yuan^{1,2} and Marlan O Scully^{1,2,3}

¹ Texas A&M University, College Station, TX 77843, USA

² Princeton University, Princeton, NJ 08544, USA

³ Baylor University, Waco, TX 76706, USA

E-mail: asvid@physics.tamu.edu

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Abstract. It has been known for two decades that coherence can yield lasing without inversion (LWI), which could be useful for making lasers at shorter wavelength. However, excitation of extreme ultraviolet and x-ray atomic transitions typically requires a plasma medium with rapid collisions, which destroy atomic coherence. Here we demonstrate LWI on a time scale shorter than the decoherence time. We show that in such a regime LWI is possible in a V -scheme with a strong coherent drive on the low-frequency transition and obtain an analytical expression for the gain of the laser pulse at high frequency. We propose an experiment in which such LWI can be realized in He plasma.

The concept of lasing without population inversion (LWI) holds promise for making lasers in the extreme ultraviolet (XUV) and x-ray spectral regions where population inversion is hard to achieve. Many schemes for LWI have been proposed in the literature [1–10]. Some of them utilize external fields to generate atomic coherence, which yields gain in the absence of population inversion via quantum interference between different transitions. For example, in the Λ -scheme shown in figure 1(a), if a – c transition is driven by a laser and spontaneous decay rate $\gamma_{a \rightarrow c} > \gamma_{a \rightarrow b}$ then coherence ρ_{ac} reaches quasi-steady state. Under such a condition ρ_{ac} can yield LWI at the a – b transition [3]. Several experiments have provided evidence of amplification and LWI in the optical domain [11].

⁴ Author to whom any correspondence should be addressed.



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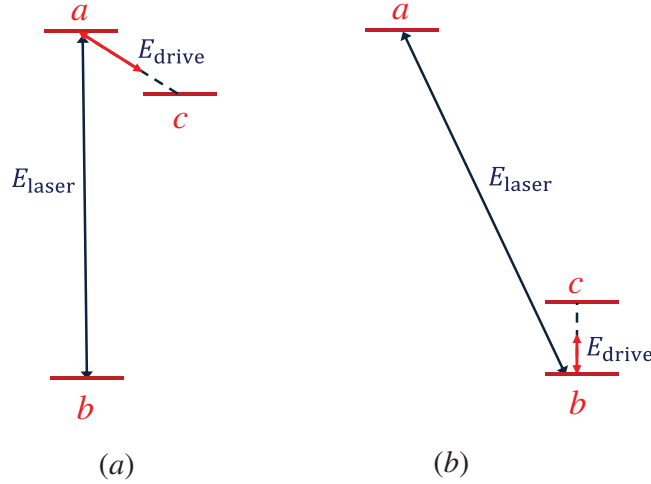


Figure 1. Energy level diagram in Λ - and V-schemes.

Previous theoretical investigations mainly studied LWI assisted by spontaneous decay or incoherent pump, which implies that their rates should be greater than collisional decoherence. However, excitation mechanisms for XUV and x-ray lasers require fast collisions challenging the possibility of having LWI in such systems.

The recombination mechanism is a promising technique for pumping XUV and x-ray lasers. In this method, a high-intensity ultrashort optical pulse ionizes atoms via tunneling. Then rapid three-body recombination and de-excitation processes follow, during which transient atomic excitation is created. This scheme is attractive due to its potential in achieving lasing at short wavelengths with relatively moderate pumping requirements. Gain and lasing with inversion have been demonstrated in such a system [12].

In a recent experiment, it has been shown that atomic coherence can yield superradiant decay of the 2^3P_1 state of He in plasma on a time scale much shorter than a collision time of 0.1 ns, whereas single atom spontaneous decay time is 100 ns [13]. Hence, coherence effects can play a role in systems with fast collisions. Motivated by this result, here we study the possibility of LWI occurring on a time scale much shorter than spontaneous decay and fast collision times. This allows us to take advantage of coherence effects, but requires investigation of the system's dynamics in a transient regime far from steady state.

Previous studies of LWI in the transient regime [10, 14–17] emphasized the importance of spontaneous decay or incoherent pumping in achieving light amplification without inversion. For example, transient LWI in a resonant V-type system has been studied in [15]. The authors found that integrated gain on the lasing transition is positive only in the presence of an incoherent pump field. Having a short time scale in mind, here we study transient LWI completely disregarding assisting factors such as spontaneous decay or incoherent pumping. In particular, we obtain an analytical expression for the gain which provides insight on LWI conditions, e.g. at what frequency the seed laser pulse is being amplified.

We consider a model in which transient amplification of a high-frequency laser field by driving a low-frequency transition is pronounced. Our model consists of an off-resonant V-scheme in which ground state b is dipole coupled with states a and c (see figure 1(b)). For simplicity of the analysis, we assume that strong field with Rabi frequency

$$\Omega^{\text{drive}}(t, z) = \Omega_d \cos(\nu_d t - k_d z) \quad (1)$$

drives only c – b transition, while the weak laser field $\Omega^{\text{laser}}(t, z)$ couples only with the a – b transition. We study how a weak seed laser field evolves in time and space. Our analysis shows that in such a scheme one can achieve gain at frequencies $\nu_{\text{laser}} = \omega_{ac} + m\nu_d$, where $m = \pm 1, \pm 3, \dots$ is an odd number, even if there is no population inversion between a and the lower levels b and c . This process can be interpreted as atomic transition from the levels a to c that is dipole forbidden with emission/absorption of an even number of photons (one laser photon is emitted and an odd number of the driving field photons is absorbed or emitted). In the present problem, evolution occurs on a time scale much faster than collision times and, hence, we can overcome atomic decoherence. Our results also remain applicable if level c lies below level b .

Under the influence of the off-resonance driving field atomic evolution is described by the following equations for the density matrix:

$$\dot{\rho}_{cb} = -i\omega_{cb}\rho_{cb} + i\Omega^{\text{drive}}(\rho_{bb} - \rho_{cc}), \quad (2)$$

$$\dot{\rho}_{cc} = i\Omega^{\text{drive}}\rho_{bc} - i\Omega^{\text{drive}*}\rho_{cb}, \quad (3)$$

$$\rho_{bb} + \rho_{cc} = \text{const}, \quad (4)$$

where $\omega_{cb} = \omega_c - \omega_b$ is the c – b transition frequency.

Applying the slowly varying envelope approximation for the laser field Ω^{laser} we obtain the following evolution equations for the slowly varying functions Ω_l , ρ_{ab}^l and ρ_{ac}^l :

$$\dot{\rho}_{ab}^l = i\Omega_l(\rho_{bb} - \rho_{aa}) - i\Omega^{\text{drive}}e^{i\omega_{cb}t}\rho_{ac}^l, \quad (5)$$

$$\dot{\rho}_{ac}^l = i(\Omega_l\rho_{bc} - \Omega^{\text{drive}*}\rho_{ab}^l)e^{i\omega_{bc}t}, \quad (6)$$

which have to be supplemented by Maxwell's equation

$$\left(c\frac{\partial}{\partial z} + \frac{\partial}{\partial t}\right)\Omega_l = i\Omega_a^2\rho_{ab}^l, \quad (7)$$

where

$$\Omega_a = \sqrt{\frac{3N\lambda_{ab}^2\gamma c}{8\pi}} \quad (8)$$

is the collective atomic frequency, N is the atomic density and γ is spontaneous decay rate of the a – b transition. Since the laser field is assumed to be weak, in equations (5) and (6) ρ_{bb} , ρ_{aa} and ρ_{bc} are determined only by the driving field and initial conditions.

Taking the time derivative of both sides of equation (7), taking $\dot{\rho}_{ab}^l$ from equation (5) and introducing

$$\tilde{\Omega}^{\text{drive}} = \Omega^{\text{drive}}e^{i\omega_{cb}t}, \quad \tilde{\rho}_{bc} = \rho_{bc}e^{-i\omega_{cb}t},$$

we find

$$\left(c\frac{\partial}{\partial z} + \frac{\partial}{\partial t}\right)\frac{\partial\Omega_l}{\partial t} + \Omega_a^2(\rho_{bb} - \rho_{aa})\Omega_l - \Omega_a^2\tilde{\Omega}^{\text{drive}}\rho_{ac}^l = 0, \quad (9)$$

where ρ_{ac}^l obeys equation

$$\dot{\rho}_{ac}^l = i\Omega_l\tilde{\rho}_{bc} - \frac{\tilde{\Omega}^{\text{drive}*}}{\Omega_a^2}\left(c\frac{\partial}{\partial z} + \frac{\partial}{\partial t}\right)\Omega_l. \quad (10)$$

Next, we solve equations (2)–(4), (9), (10) and find laser modes that grow exponentially with time. We assume that at $t = 0$ there is some population at the levels a and b , but no population at the level c and no initial coherence. The driving field is turned on adiabatically. We assume that under the influence of the driving field populations of the levels b and c undergo small changes. Then equations (2)–(4) yield

$$\tilde{\rho}_{bc} = -\frac{\Omega_d}{2} \left(\frac{e^{i\nu_d t - ik_d z}}{\nu_d - \omega_{cb}} - \frac{e^{-i\nu_d t + ik_d z}}{\nu_d + \omega_{cb}} \right) e^{-i\omega_{cb} t} \rho_{bb}(0), \quad (11)$$

$$\rho_{bb} = \rho_{bb}(0) - \delta [1 - \cos(2\nu_d t - 2k_d z)], \quad (12)$$

where

$$\delta = \frac{\Omega_d^2 \rho_{bb}(0)}{2(\nu_d^2 - \omega_{cb}^2)} \ll 1. \quad (13)$$

Next, we write ρ_{ac}^1 as

$$\rho_{ac}^1 = e^{i(\nu_d - \omega_{cb})t - ik_d z} \rho_1 + e^{-i(\nu_d + \omega_{cb})t + ik_d z} \rho_2, \quad (14)$$

which gives the following equations for Ω_1 , ρ_1 and ρ_2 :

$$\left(c \frac{\partial}{\partial z} + \frac{\partial}{\partial t} \right) \frac{\partial \Omega_1}{\partial t} + \Omega_a^2 [\rho_{bb}(0) - \rho_{aa} + \delta \cos(2\nu_d t - 2k_d z)] \Omega_1 - \frac{\Omega_d \Omega_a^2}{2} (\rho_1 + \rho_2 + e^{2i\nu_d t - 2ik_d z} \rho_1 + e^{-2i\nu_d t + 2ik_d z} \rho_2) = 0, \quad (15)$$

$$\dot{\rho}_1 + i(\nu_d - \omega_{cb})\rho_1 = -\frac{\Omega_d}{2} \left[\frac{1}{\Omega_a^2} \left(c \frac{\partial}{\partial z} + \frac{\partial}{\partial t} \right) + \frac{i\rho_{bb}(0)}{\nu_d - \omega_{cb}} \right] \Omega_1, \quad (16)$$

$$\dot{\rho}_2 - i(\nu_d + \omega_{cb})\rho_2 = -\frac{\Omega_d}{2} \left[\frac{1}{\Omega_a^2} \left(c \frac{\partial}{\partial z} + \frac{\partial}{\partial t} \right) - \frac{i\rho_{bb}(0)}{\nu_d + \omega_{cb}} \right] \Omega_1. \quad (17)$$

We look for a solution for Ω_1 in the form

$$\Omega_1 = e^{ikz - i\nu t}, \quad (18)$$

where ν is detuning of Ω_1 from the transition frequency ω_{ab} . We assume that k is real. Then the imaginary part of ν gives gain G (absorption) per unit time of the mode with wavenumber k . During propagation of the seed laser pulse Ω_l through the medium it grows as $\exp(Gt)$, where $G = \text{Im}(\nu)$.

Substituting equation (18) into equations (16) and (17) gives

$$\rho_1 = -\frac{\Omega_d}{2(\nu_d - \omega_{cb} - \nu)} \left[\frac{1}{\Omega_a^2} (ck - \nu) + \frac{\rho_{bb}(0)}{\nu_d - \omega_{cb}} \right] e^{ikz - i\nu t}, \quad (19)$$

$$\rho_2 = \frac{\Omega_d}{2(\nu_d + \omega_{cb} + \nu)} \left[\frac{1}{\Omega_a^2} (ck - \nu) - \frac{\rho_{bb}(0)}{\nu_d + \omega_{cb}} \right] e^{ikz - i\nu t}. \quad (20)$$

Plugging equations (18)–(20) into equation (15) and disregarding terms oscillating with frequency $2\nu_d$ yields the following equation for ν :

$$\nu (ck - \nu) + \Omega_a^2 (\rho_{bb}(0) - \rho_{aa}) - \frac{\Omega_d^2 \Omega_a^2}{4} \left(\frac{1}{(\nu + \nu_d + \omega_{cb})} \left[\frac{ck - \nu}{\Omega_a^2} - \frac{\rho_{bb}(0)}{\nu_d + \omega_{cb}} \right] + \frac{1}{(\nu - \nu_d + \omega_{cb})} \left[\frac{ck - \nu}{\Omega_a^2} + \frac{\rho_{bb}(0)}{\nu_d - \omega_{cb}} \right] \right) = 0. \quad (21)$$

This equation has resonant character, namely, when $\nu = \pm \nu_d - \omega_{cb}$ the last terms become large. This is the region of maximum gain. The optimum value of k is obtained from the condition that one of the roots of the quadratic equation

$$\nu (ck - \nu) + \Omega_a^2 (\rho_{bb}(0) - \rho_{aa}) = 0$$

is equal to $\nu = \pm \nu_d - \omega_{cb}$, which gives

$$ck = \pm \nu_d - \omega_{cb} + \frac{\Omega_a^2 (\rho_{bb}(0) - \rho_{aa})}{\omega_{cb} \mp \nu_d}. \quad (22)$$

Plugging this in equation (21) yields the final expression for the maximum gain per unit time

$$G = \frac{\Omega_d \Omega_a \sqrt{\rho_{aa}}}{2\sqrt{(\nu_d \mp \omega_{cb})^2 + \Omega_a^2 (\rho_{bb}(0) - \rho_{aa})}}. \quad (23)$$

Equation (23) is valid provided that G is much larger than decoherence rate γ_{tot} . Laser light is emitted at frequencies $\nu_{\text{laser}} = \nu + \omega_{ab} = \omega_{ac} \pm \nu_d$.

Gain per unit length G_L can be obtained in a similar way. Now one should treat ν as real and introduce decoherence γ_{tot} in equation (21). Then imaginary part of k gives gain (absorption) per unit length G_L of the mode with frequency ν : $G_L = -\text{Im}(k)$. The gain is maximum near resonance when $\nu \approx \pm \nu_d - \omega_{cb}$, which gives

$$G_L = -\text{Im}(k) = \frac{3N\lambda_{ab}^2 \gamma_{\text{tot}} [\rho_{aa} \Omega_d^2 - 4\gamma_{\text{tot}}^2 (\rho_{bb}(0) - \rho_{aa})]}{32\pi \gamma_{\text{tot}} (\nu_d \mp \omega_{cb})^2 + \gamma_{\text{tot}}^2 + \Omega_d^2/2}. \quad (24)$$

Equation (24) shows that there is gain if

$$\Omega_d^2 > 4\gamma_{\text{tot}}^2 \left(\frac{\rho_{bb}(0)}{\rho_{aa}} - 1 \right), \quad (25)$$

that is, to achieve LWI, strength of the driving field Ω_d must exceed decoherence rate.

If levels b and c are degenerate then equations (2)–(4) have exact solutions for any strength of the driving field

$$\rho_{bb}(t) = \rho_{bb}(0) \cos^2 \left[\frac{\Omega_d}{\nu_d} \sin(\nu_d t - k_d z) \right], \quad (26)$$

$$\rho_{bc}(t) = -\frac{i}{2} \rho_{bb}(0) \sin \left[\frac{2\Omega_d}{\nu_d} \sin(\nu_d t - k_d z) \right], \quad (27)$$

where $\rho_{bb}(0)$ is the initial population of the level b and $\rho_{cc}(0) = 0$.

Next we solve equations (9) and (10) with Ω^{drive} , $\rho_{bb}(t)$ and $\rho_{bc}(t)$ given by equations (1), (26), (27) and $k_d = 0$ numerically and compare the results with our analytical findings. We look for solution in the form

$$\Omega_l(t, z) = e^{ikz} \Omega_l(t), \quad \rho_{ac}^l(t, z) = e^{ikz} \rho_{ac}^l(t),$$

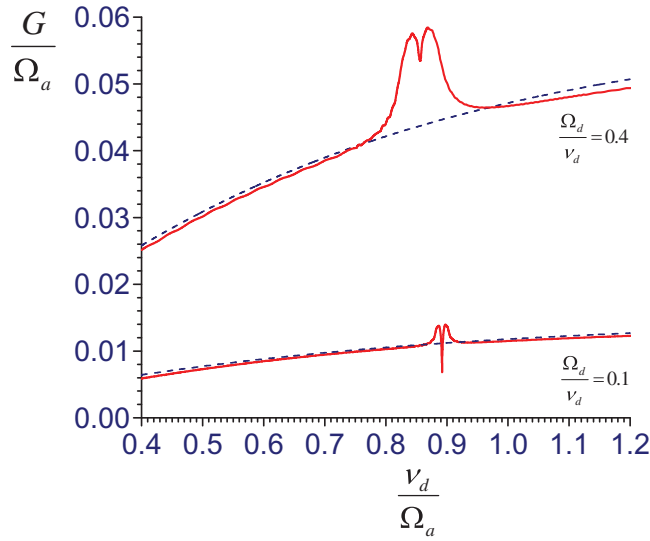


Figure 2. Maximum gain as a function of the driving field frequency ν_d in the V-scheme. Initial population of the ground state is $\rho_{bb}(0) = 0.9$ and $\rho_{aa} = 0.1$. Driving field strength is $\Omega_d/\nu_d = 0.4$ and 0.1 . Plot is obtained by numerical solution of equations (9) and (10). Analytical result (23) is shown by the dashed line.

where k is a real number. We found that if $\rho_{aa} \neq 0$, then for certain k laser field Ω_l exponentially grows even if there is no population inversion between levels a and b .

Figure 2 shows gain per unit time G on the lasing transition as a function of the driving field frequency ν_d for optimum value of k , which maximizes G . Initial populations are $\rho_{bb} = 0.9$, $\rho_{aa} = 0.1$ and $\rho_{ab} = 0$. Driving field strength is $\Omega_d/\nu_d = 0.4$ and 0.1 . For such parameters there is no population inversion between levels a and b at any moment of time. Figure 2 demonstrates that there is large gain in a broad range of the driving field frequencies. We plot the analytical result (23) as dashed lines in figure 2. Features in the vicinity of $\nu_d = \Omega_a \sqrt{\rho_{bb}(0) - \rho_{aa}} = 0.89\Omega_a$ are a manifestation of collective parametric resonance, which is beyond our analytical treatment and will be further developed elsewhere.

To demonstrate that the seed laser pulse gains its energy from the atomic population of level a (and not from the driving field) we solve the full system of Maxwell–Schrödinger equations numerically. We initially take $\rho_{bb} = 0.9$, $\rho_{aa} = 0.1$ and $\rho_{ab} = 0$. In simulations, we assume that levels b and c are degenerate and the driving field is given by equation (1) with $\Omega_d = 0.3\nu_d$ and $k_d = \nu_d/c$. We send a weak laser pulse of Gaussian shape and duration 1.6 ps into atomic sample of length $L = 0.3$ cm and calculate how pulse energy W^{laser} evolves with time. Strength of the a – b transition and atomic density are chosen such that collective atomic frequency is $\Omega_a = 10^{13} \text{ s}^{-1}$ (which corresponds to atomic density $N \sim 10^{18} \text{ cm}^{-3}$). Figure 3 shows $W^{\text{laser}}/W_0^{\text{atom}}$ as a function of time (lower curve). Here W_0^{atom} is the initial energy stored in atomic excitation. The upper curve shows average (over atomic sample) population of the excited state $\rho_{aa}(t)$. For short evolution time the laser pulse grows exponentially but remains weak to affect ρ_{aa} . This is the linear gain regime. Later on pulse energy starts to saturate and population ρ_{aa} gets depleted. The sum of two curves (the net energy of atoms and field) remains constant, which is shown as a dashed line in figure 3. This implies that laser pulse energy grows at the expense of ρ_{aa} .

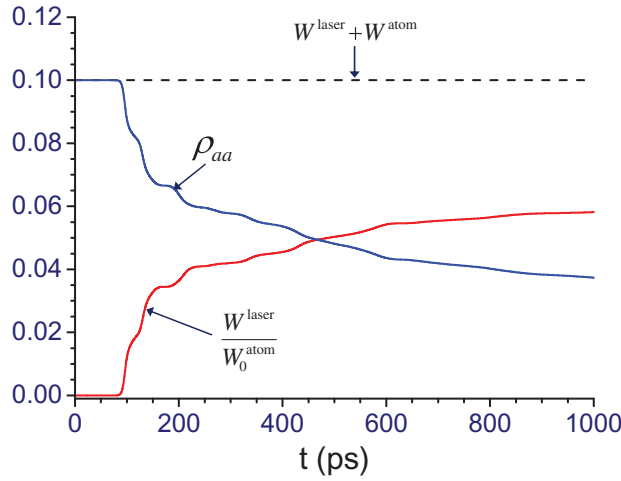


Figure 3. Energy of the seed laser pulse W^{laser} (lower curve) and average population ρ_{aa} of the level a (upper curve) as a function of time obtained by numerical solution of the Maxwell–Schrödinger equations with initial conditions $\rho_{bb} = 0.9$, $\rho_{aa} = 0.1$ and $\rho_{ab} = 0$. Degenerate levels b and c are driven by coherent field (1) with $\Omega_d = 0.3\nu_d$ and $k_d = \nu_d/c$. The dashed line is the sum of two curves.

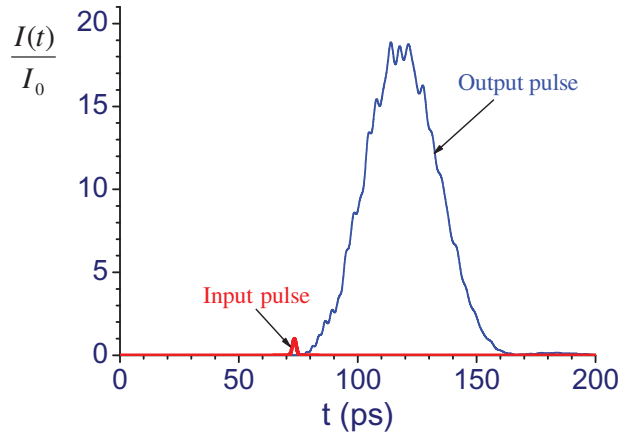


Figure 4. Intensity $I(t)$ of the input and output UV laser pulse after it propagates through 1 cm of He gas driven by an IR coherent field.

We also considered the Λ -scheme shown in figure 1(a). We found that for this configuration, unlike the V-scheme, there is no gain without inversion in the transient regime.

Next we discuss a possible experiment to demonstrate transient LWI at a high-frequency transition produced by driving a lower frequency transition. Active medium could be, for example, a gas of He atoms partially excited in the metastable triplet 2^3S_1 state (level b in the present notations) inside helium plasma, as in the recent experiment [13]. One can drive an infrared $2^3P_1 \rightarrow 2^3S_1$ (1083 nm) transition and generate lasing at the UV $3^3P_1 \rightarrow 2^3S_1$ (388.9 nm, $\gamma = 10^7 \text{ s}^{-1}$) frequency without population inversion. For density of atoms in the metastable state $N = 10^{16} \text{ cm}^{-3}$ collective atomic frequency for the lasing transition is $\Omega_a = 7.54 \times 10^{11} \text{ s}^{-1}$.

We assume that initially $\rho_{bb} = 0.85$, $\rho_{aa} = 0.15$ and $\rho_{cc} = 0$. If the IR transition is driven by a laser with Rabi frequency $\Omega_d = 6 \times 10^{11} \text{ s}^{-1}$ detuned by $\Delta = \Omega_d$ from the resonance and decoherence rate is $\gamma_{\text{tot}} = 10^{10} \text{ s}^{-1}$, then equation (23) yields that gain of the UV laser pulse per unit time is $G = 10^{11} \text{ s}^{-1} \gg \gamma_{\text{tot}}$, while equation (24) predicts that gain per unit length is $G_L = 47 \text{ cm}^{-1}$. As a demonstration, we solve the Maxwell–Schrödinger equations for the evolution of a weak Gaussian laser pulse with duration 1.6 ps numerically for the present parameters. Figure 4 shows intensity $I(t)$ of the input and output UV laser pulse after it propagates through the medium of length $L = 1 \text{ cm}$. Intensity is normalized by the peak intensity of the input pulse I_0 . The plot indicates that pulse energy increases by several orders of magnitude.

In summary, we show that LWI can be achieved in systems with fast collisional decoherence on a short time scale, which utilizes coherence effects. This is relevant to experiments on XUV and x-ray lasers in which transient atomic population is created by ionization–recombination processes. In particular, we find that transient LWI is possible in the V-scheme in which the low-frequency transition is coherently driven by a field with Rabi frequency exceeding the decoherence rate. This is, however, not the case in the Λ -scheme that gives no transient gain. We found an analytical expression for the gain that yields insight on LWI conditions and discuss an experiment where transient LWI can be realized.

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